PART 1

INTRODUCTION

Chapter 2. PCBs Modeling Overview

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1.2.1 Background

The mass balance project was based upon the Enhanced Monitoring Program (EMP), a comprehensive, two-year synoptic survey for selected toxic chemicals in the Lake Michigan ecosystem. The EMP included tributary load and atmospheric deposition monitoring; ambient water column, biota, and sediment sampling; and additional measurements to define and confirm transport and fate processes. The toxics studied for the Lake Michigan Mass Balance Project (LMMBP) included polychlorinated biphenyls (PCBs), atrazine, transnonachlor, and mercury. The project was led by the United States Environmental Protection Agency

(USEPA)/Great Lakes National Program Office (GLNPO). Modeling support to the project was provided by the USEPA/Mid-Continent Ecology Division (MED)/Office of Research and Development (ORD)/Large Lakes Research Station (LLRS) in cooperation with the Atmospheric Research and Exposure Assessment Laboratory (AREAL); the National Oceanic and Atmospheric Administration (NOAA)/Great Lakes Environmental Research Laboratory (GLERL); and other cooperators. The research developed a suite of integrated mass balance models to simulate the transport, fate, and bioaccumulation of toxic chemicals in Lake Michigan.

1.2.2 Modeling Objectives

Development of effective strategies for toxics management requires a quantitative understanding of the relationships between sources, inventories. concentrations, and effects of contaminants in the ecosystem. A mass balance modeling approach was used to address the relationship between sources of toxic chemicals and concentrations in air, water, sediment, and biota. This approach integrated load estimation, ambient monitoring, and research efforts within a modeling framework that was compatible with both scientific as well as ecosystem management objectives. The mass balance approach estimated the magnitude of mass fluxes that constitute the pathways for toxics transport into and out of the lake, that distribute toxics within the lake water column and sediment, and that lead to bioaccumulation of the aquatic food webs. Based upon these estimates, the mass balance was used to determine the rate of change in concentrations and inventories of toxics as inputs such as atmospheric

and tributary loadings change. Thus, the mass balance can serve as a useful tool to estimate or predict the outcome of alternatives under consideration for toxics management.

Modeling efforts associated with the LMMBP had the following objectives:

- 1. Provide a consistent framework for integrating load estimates, ambient monitoring data, process research efforts, and prior modeling efforts, leading to a better understanding of toxic chemical sources, transport, fate, and bioaccumulation in Lake Michigan.
- 2. Estimate the loading of priority toxics, solids, and nutrients from all major tributaries to Lake Michigan for the duration of the study.
- 3. Estimate the atmospheric deposition and airwater exchange of priority toxics, including spatial and temporal variability over Lake Michigan.
- 4. Calibrate and confirm mass balance models for priority toxics using project data, based upon models for hydrodynamic and sediment transport, eutrophication/organic carbon dynamics, toxics transport and fate, and food web bioaccumulation.
- 5. Based upon the mass balance models, evaluate the magnitude and variability of toxic chemical fluxes within and between lake compartments, especially between the sediment and water column and between the water column and the atmosphere.
- Apply the calibrated mass balance models to forecast contaminant concentrations in water and sediment throughout Lake Michigan, based upon meteorological forcing functions and future loadings based upon load reduction alternatives.
- 7. Predict the bioaccumulation of persistent toxic chemicals through the food web leading to top predator fish (lake trout and coho salmon) for location-specific fish populations in the lake, in order to relate mass balance predictions of water and sediment exposure to this significant impaired use.

- 8. Estimate (quantify) the uncertainty associated with estimates of tributary and atmospheric loads of priority toxics, and model predictions of contaminant concentrations.
- Identify and prioritize further monitoring, modeling, and research efforts to (1) address additional toxic substances, (2) further reduce uncertainty and improve accuracy of predictions, (3) establish additional cause-effect linkages, such as ecological risk endpoints and feedbacks, and (4) evaluate additional source categories, such as non-point sources in the watershed.

The purpose of PCBs modeling was to simulate their transport, fate, and bioaccumulation in Lake Michigan. PCBs are a group of persistent, bioaccumulative hydrophobic organic chemicals (HOCs) that are ubiquitous in the Great Lakes. Although anthropogenic inputs from production and disposal largely ceased following their ban in the 1970s, atmospheric and watershed tributary transport pathways to the lake continue the import of PCBs. In addition, a large in-lake sediment inventory represents an internal source of PCBs, which are recycled annually. PCBs have been consistently identified as the contaminant of greatest concern to human and ecosystem health in the Great Lakes (Ludwig et al., 1993; Swain, 1991; Gilbertson et al., 1991).

1.2.3 Historical Modeling

The modeling design and approach for the LMMBP reflects a progression of prior modeling efforts in Lake Michigan and throughout the Great Lakes. These include eutrophication and toxic substance mass balance models, food web bioaccumulation models, and predictive hydrodynamic and sediment transport models. Although not a comprehensive review, several of these prior modeling efforts are discussed below.

1.2.3.1 Lake-1

A eutrophication model for Lake Michigan was developed by Rodgers and Salisbury (1981), based upon the Lake-1 model which was also applied to Lakes Erie, Huron, and Ontario. The model was calibrated and tested using data from 1976 and 1977.

The importance of climatic factors on limnological (including eutrophication) processes in Lake Michigan was demonstrated, as the severe winter and extensive ice cover of 1976-1977 dramatically reduced total phosphorus concentrations in the second year. This work also identified several refinements necessary for accurate modeling of eutrophication: phosphorus availability to phytoplankton and particle transport including shoreline erosion and sediment resuspension were apparently significant influences upon nutrient and phytoplankton dynamics observed in Lake Michigan.

1.2.3.2 Completely-Mixed Model

A lakes-in-series model for conservative substances was developed by Sonzogni *et al.* (1983) and applied to forecast chloride concentrations in each of the Great Lakes as a function of expected future loadings. This model demonstrated that concentrations of non-reactive substances would substantially "lag" the history of their input. This was especially the case for Lake Michigan, where maximum chloride concentrations were not predicted to occur until the 22nd Century despite declining loads after the 1970s. Similarly strong, non-steady-state behavior may be expected for other chemicals which are non-reactive and weakly associated to particles.

1.2.3.3 General Mass Balance Framework for Toxic Chemicals in the Great Lakes

At about the same time, models were being developed which would serve as the foundation for describing and simulating the transport and fate of hydrophobic chemicals in the Great Lakes. Thomann and Di Toro (1983) and Robbins (1985) demonstrated that the lake-wide, annual concentration trend of contaminants including cesium-137, plutonium-239/240, and PCBs, were dependent upon particle transport between the water column and a resuspendable sediment compartment. The principal loss mechanisms from the lakes were found to be burial by sedimentation and (for PCBs) volatilization. The somewhat paradoxical behavior of these models was that the water column contaminant dynamics were largely controlled by sediment parameters.

1.2.3.4 Food Web Bioaccumulation Model

A food web bioaccumulation model was developed by Thomann and Connolly (1984) and applied to simulate bioaccumulation of PCBs in Lake Michigan The model was confirmed with an lake trout. extensive data set collected in 1971, including nine age classes of trout, diet characterization by gut contents analysis, and alewife. The model was successful in predicting bioaccumulation for mature age classes of lake trout, although not for juveniles. Dietary transfer was demonstrated to be the predominant route of PCBs accumulation, in comparison to direct chemical uptake from water. Substantial residual variance in lake trout PCBs concentrations within age class CV = 1 was not explained by this lake-wide, average-individual model.

1.2.3.5 MICHTOX

An integrated mass balance and bioaccumulation model for PCBs (modeled as two homologs) and 10 other toxic chemicals was developed as a planning tool for the LMMBP (Endicott et al., 2005). The MICHTOX mass balance was calibrated to suspended solids and plutonium data for the southern lake basin, while the bioaccumulation model combined Thomann and Connolly's (1984) effort with chemical-specific parameterization from Lake Ontario. MICHTOX demonstrated that reasonable predictions of PCB concentration trends in water. sediment, and biota could be developed although significant uncertainties regarding sediment-water and air-water contaminant transport remained. These are the most significant transport fluxes for PCBs and presumably other hydrophobic contaminants. Major data gaps for other priority toxics allowed only order-of-magnitude estimates of load-concentration relationships. When this model was developed and run, available monitoring data for toxic chemical concentration in tributaries, air, lake water, sediment, and biota were not adequate to define loading trends or to relate the distribution of loadings to contaminant gradients observed for sediment and biota. Credible model predictions of toxic chemical transport, fate, and bioaccumulation would depend upon developing a comprehensive data set quantifying loadings, sediment inventories,

concentrations, and transport fluxes on a spatiallyresolved basis, and localized descriptions of food web structures.

1.2.3.6 Green Bay Mass Balance Project

The Green Bay Mass Balance Project (GBMBP) demonstrated the feasibility of applying mass balance principles to manage toxic chemicals in the Great Lakes ecosystem. A two-year (1989-1990) synoptic sampling program was designed to collect appropriate and complete data for the mass balance study. A suite of integrated mass balance and bioaccumulation models were developed which, together, provided an ecosystem-level simulation of sources, transport, fate, and bioaccumulation of PCBs throughout the Fox River and Green Bay. This study advanced the state-of-the-art of mass balance modeling, particularly the ability to construct a fairly complete and accurate description of contaminant mass transport.

Several aspects of the Green Bay modeling effort were noteworthy. Particle transport and sorption processes were found to be of fundamental importance as bases for contaminant modeling. Resuspension of contaminated sediments in the Fox River constituted the major source of PCBs to the river as well as the bay. In the bay, particle sorbent dynamics were strongly affected by phytoplankton production and decay. The relative significance of hydraulic transport, sediment transport, burial, volatilization, and open lake boundary exchange processes upon the PCBs mass balance varied considerably with location in Green Radionuclide tracers were again essential for calibration of particle fluxes and confirmation of longterm contaminant transport predictions. significance of contaminant accumulation at the base of the food web, and fish movement in relation to exposure gradients, were demonstrated in the bioaccumulation model. The LMMBP demonstrated the linked submodel approach to ecosystem model development and application, and the feasibility of using such a model for assessing the effectiveness of toxics management control alternatives.

1.2.3.7 SEDZL

The Green Bay Mass Balance Project (GBMBP) also provided data to test a predictive two-dimensional, hydrodynamic and sediment transport model of the Fox River, SEDZL (Gailani et al., 1991). SEDZL incorporates realistic descriptions of cohesive sediment resuspension, flocculation, and deposition processes, and contaminant sorption, which are critical for accurate prediction of hydrophobic contaminant transport. These process descriptions were based on laboratory and field experiments with river, bay, and lake sediments. A three-dimensional bed submodel was used to describe sediment bed properties which varied with depth as well as location. The fine spatial resolution of the model allowed detailed simulation of in-place pollutant transport in both the water column and sediment bed. Although computationally intensive and requiring specialized data, SEDZL has substantially advanced the state-of-the-art for sediment and contaminant transport modeling in the Great Lakes. SEDZL had also been applied to the Buffalo and Saginaw Rivers as part of the ARCS/RAM project (Gailani et al., 1994: Cardenas and Lick, 1996). These applications included long-term forecasts (10-25 years) of sediment and contaminant transport. SEDZL had also been applied to large water bodies such as Lake Erie, and marine coastal waters including Santa Barbara Channel, and Atchafalaya Bay (Lick et al., 1994; Pickens, 1992) where wave action as well as currents force sediment resuspension.

1.2.4 Model Resolution

Model resolution is the spatial and temporal scale of predictions, as well as the definitions of model state variables. While factors such as data availability, model sophistication, and computer resources constrain resolution to a degree, different levels of model resolution are possible and, are in fact, necessary. Three "levels" of spatial resolution, indicated by the segmentation grid of the lake surface, are illustrated in Figure 1.2.1. Level 1 was resolved at the scale of lake basins (characteristic length, L = 150 km), with an associated seasonal temporal resolution. This was a screening-level model resolution used in MICHTOX. Level 2 was

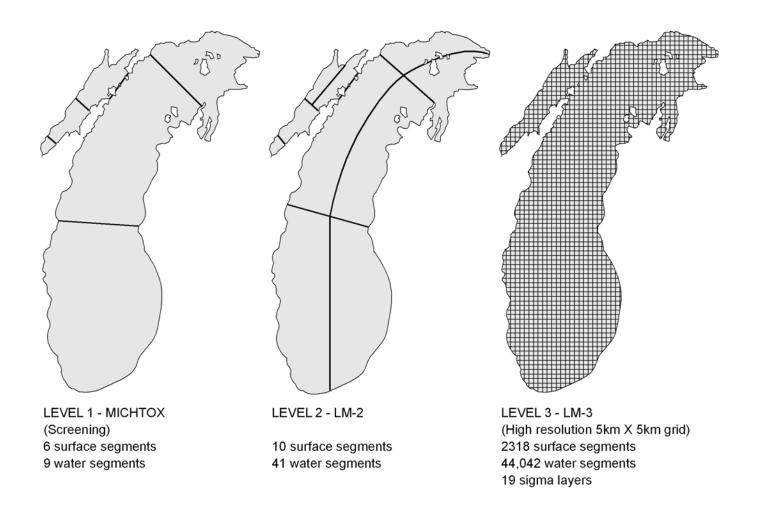


Figure 1.2.1. Surface water segmentation for alternative Lake Michigan mass balance model levels.

resolved at a regional scale defined by food webs (L = 40 km) including gross resolution of the nearshore and offshore regions; temporal resolution was weekly-to-monthly. This resolution was roughly comparable to that achieved by models developed in the GBMBP. Level 3 was a hydrodynamic scale resolution (L = 5 km), with associated daily temporal resolution. Level 3 was scaled to resolve and predict particle transport processes as well as hydrodynamic transport.

Although the Lake-wide Management Plan (LaMP) and the Great Waters Program objectives are "lake-wide," both of these emphasize biotic impairments occurring primarily in localized, nearshore regions. LaMP objectives also require that the transport of contaminants from tributaries and other nearshore sources to the open lake be resolved. Therefore, the

Level 1 model was not adequate for the study objectives. Level 2 resolution was adequate for most modeling objectives, but not for resolution of significant hydrodynamic and sediment transport events. Level 3 resolution was required for accurate hydrodynamic and sediment transport modeling and was desirable for predicting nearshore gradients, especially those formed by transients such as thermal bars, upwelling, and storm-induced resuspension; as well as more persistent features such as tributary plumes, thermal stratification, and the benthic nepheloid layer. Level 3 transport resolution was also valuable in relating toxics loading from the 10 Areas of Concern (AOCs) adjoining Lake Michigan, which must be addressed by the Remedial Action Plan (RAP) process, to the LaMP via the LMMBP.

The modeling design for the LMMBP was based on the development of a number of models at three levels of resolution. For the contaminant transport and fate (CTF) models, MICHTOX was resolved at Level 1, and LM2-Toxic was resolved at Level 2. For the eutrophication models, MICH1 was resolved at Level 1 and LM3-Eutro was resolved at Level 3. The Princeton Ocean Model (POM) and atmospheric loading models were resolved at Level 3. Results of the hydrodynamic model were spatially and temporally averaged prior to coupling to the Level 2 The rationale for specifying different resolutions was the hydrodynamic models require a Level 3 resolution to offer the best capability for transport simulation and forecasting. resolution was specified for LM2-Toxic because this model had been demonstrated at this resolution.

1.2.5 Models Developed and Applied

The model design for the LMMBP was based upon the linked submodel approach used in the GBMBP. Models developed, refined, and applied by the Large Lakes and Rivers Forecasting Research Branch (LLRFRB) included eutrophication/sorbent dynamics (MICH1 and LM3-Eutro), contaminant transport and fate (MICHTOX and LM2-Toxic), and food web bioaccumulation (LM Food Chain) models (Figure 1.2.2). Models developed and run elsewhere included a hydrodynamics model (POM) (Schwab and Beletsky, 1998), an atmospheric loading model (Green et al., 2000; Miller et al., 2001), and a tributary loading model (Hall and Robertson, 1998). Only the models developed, refined, and applied at LLRFRB will be discussed in detail within this document.

1.2.5.1 Lake Process Models

The mass balance for toxics in Lake Michigan was comprised of linked hydrodynamic (POM), eutrophication/sorbent dynamics (LM3-Eutro), contaminant transport and transformation (LM2-Toxic), and bioaccumulation simulations (LM Food Chain). In addition, Level 1 eutrophication/sorbent dynamics (MICH1) and contaminant transport and transformation/bioaccumulation (MICHTOX) simulations were run for comparison to Level 3 and 2 results, respectively. Each of these models represented significant processes affecting the mass balance for toxic chemicals. The hydrodynamic

model predicted water movements necessary to describe the three-dimensional transport of dissolved and particulate constituents in the water column. The eutrophication models described the production, respiration, grazing, and decomposition of planktonic biomass within the lake. The contaminant transport and fate models described contaminant partitioning between dissolved and sorbed phases, mass transfer (air, water, between media sediment). biogeochemical transformations. bioaccumulation models simulated contaminant accumulation from water and sediments to predator fish via direct exposure and trophic transfer through benthic and pelagic food webs. Together, these submodels formed an integrated description of toxic chemical cycling in the aquatic ecosystem with which to predict the relationship between loadings and concentrations of PCBs.

1.2.5.2 Hydrodynamics (POM)

The Princeton Ocean Model (POM) (Blumberg and Mellor, 1980, 1987) was used to compute threedimensional current fields in the lake. The POM simulated large- and medium (km)-scale circulation patterns, vertical stratification and velocity distribution, seiche, and surface waves. This model was also used to simulate a thermal balance for the lake. The POM is a primitive equation, numerical hydrodynamic circulation model that predicts threedimensional water column transport in response to wind stress, temperature, barometric pressure, and Coriolis force. The POM has been demonstrated to accurately simulate the predominant physics of large water bodies (Blumberg and Mellor, 1983, 1985; Blumberg and Goodrich, 1990). This model was used to develop year-long simulations on a 5 km horizontal grid, with nine sigma vertical layers, at one-hour intervals for Lake Michigan (Schwab and Observed and simulated Beletsky, 1998). meteorological data were used to define model forcing functions. Extensive measurements of temperature and current distributions collected in Lake Michigan during 1982-1983 were used to provide the necessary data for model calibration; measurements of daily surface temperature and current distributions were used to confirm hydrodynamic simulations for 1994-1995.

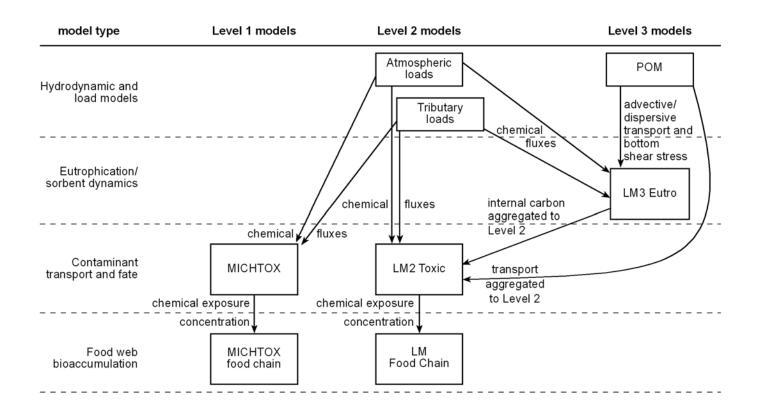


Figure 1.2.2. Model construct used for the LMMBP to model PCBs.

1.2.5.3 Eutrophication/Sorbent Dynamics (LM3-Eutro)

The eutrophication/sorbent dynamics (LM3-Eutro) model predicted the production, transformation, and decay of plankton biomass in response to seasonal dynamics of temperature, light, and nutrient concentrations. In the open lake, living and dead plankton comprise the majority of suspended particles and generate significant autochthonous loads of particulate and dissolved organic carbon (POC and DOC) to which PCBs and other contaminants preferentially partition (Richardson et al., 1983; DePinto et al., 1993). LM3-Eutro simulated the non-conservative, seasonally-variable dynamics of the biotic organic pool, which has a significant influence upon partitioning of HOCs (Dean et al., 1993). A similar, less resolute model was applied to simulate the dynamics of organic carbon states in Green Bay as part of the GBMBP (DePinto et al., 1993). Model outputs included autochthonous solids

loads and transformation and decay rates that were used as inputs for LM2-Toxic.

1.2.5.4 Contaminant Transport and Fate (LM2-Toxic)

The mass balance for toxic chemicals in the lake was computed in a contaminant transport and fate model (LM2-Toxic) which described contaminant transport, intermedia exchange, phase distribution, and biogeochemical transformations in both the water column and sediments. LM2-Toxic was calibrated and confirmed for selected individual congeners and the sum of PCBs congeners. Mass balance analyses were performed for total PCBs to evaluate the significant source, transport, and loss pathways. The effectiveness of alternative load reduction scenarios upon reducing total PCB concentrations were forecast.

1.2.5.5 Food Web Bioaccumulation (LM Food Chain)

A bioaccumulation model simulated chemical accumulation in the food web in response to chemical exposure, based upon chemical mass balances for aquatic biota. The general form of the bioaccumulation equation was well-defined, and equated the rate of change in chemical concentration within a fish (or other aquatic organism) to the sum of chemical fluxes into and out of the animal. These fluxes included direct uptake of chemical from water. the flux of chemical into the animal through feeding. and the loss of chemical due to elimination (desorption and excretion) and dilution due to growth. To predict bioaccumulation for top predator fish, the bioaccumulation mass balance was repeatedly applied to animals at each trophic level to simulate chemical biomagnification from primary and secondary producers, through forage species to top predators. Food web bioaccumulation models have been successfully applied for PCBs and other HOCs in several large-scale aquatic ecosystems (Thomann and Connolly, 1984; Connolly and Tonelli, 1985) and. most recently, for the GBMBP (Connolly et al., 1992). The model developed for that project, FDCHN, was adapted for use in Lake Michigan (LM Food Chain). FDCHN is a time-variable, population-based age class model, incorporating realistic descriptions of bioenergetic, trophodynamic, and toxicokinetic processes. The general features of FDCHN were well-suited to a modeling application such as the LMMBP. For Lake Michigan, bioaccumulation of PCB congeners was modeled for lake trout and coho salmon food webs. Food web bioaccumulation was simulated for sub-populations of lake trout in three distinct biotic zones.

1.2.6 Model Quality Assurance

A Quality Assurance Project Plan (QAPP) was prepared and implemented for the PCBs modeling (Richardson *et al.*, 2004). The QAPP specified procedures for code development, testing, modification, and documentation; as well as methods and measures applied in model calibration, confirmation, and uncertainty analysis.

1.2.7 Model Application and Computational Aspects

1.2.7.1 Annual Simulations

Annual simulations were run with the models for the period of 1994-1995. Results were analyzed in terms of regional and lake-wide contaminant loads, fluxes and inventories, and spatial and temporal gradients of contaminant concentrations. Bioaccumulation simulations were analyzed in terms of relative accumulation pathways, spatial and temporal variability of contaminant concentration ratios (bioconcentration factor, bioaccumulation factor, biota/sediment accumulation factor, predator/prey), and influence of diet, age, and migration factors.

1.2.7.2 Long-Term Simulations

Long-term simulations were used to forecast the impact of various management scenarios. Forecasts were performed to determine time to steady-state for both continuing and discontinued loads. Forecasts were also run to evaluate reductions in exposure concentrations resulting from elimination of tributary and/or atmospheric loading. These forecasts were propagated through the food web bioaccumulation model for PCBs to estimate time for sport fish contaminant concentrations to decline below criteria limits.

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